Demonstration of the quantum mechanics applicability limits and solution of the irreversibility problem

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Abstract

An experimental study of the applicability of mechanics equations to describing the process of equilibrium establishing in an isolated spin system was performed. The time-reversion effects were used at the experiments. It was demonstrated, that the equations of mechanics do not describe the experimental results. The demonstration of the incomleteness of quantum mechanics description of the macrosystems annuls the contradiction which lays in the basis of the irreversibility problem.

1 Introduction

The problem of correlation between reversibility and irreversibility in the evolution of physical systems is one of the fundamental problems of contemporary science. It is basically the problem of the unsolved contradiction between the determined world that we get if we absolutize the mechanics equations , and the real world as we know it. At the level of the law of physics, this contradiction appears in the incompatibility of the 2nd law of thermodynamics and the reversibility of physical systems evolution followed from classical and quantum mechanics equations.

In a numerous attempts to arrive to the irreversibility from the reversible equations there were an avowed or hidden assumptions that would allow the authors to arrive to a desirable result but would not have grounds within the mechanical theory.

The reversibility of the evolution of those systems that are described by the mechanics equations has given birth to the idea that the source of the irreversibility is the interaction of the system with the surrounding environment where there are statistical laws in operation. The first thing that comes to mind when one reads the works proclaiming this point of view is the question of where those statistical laws in the physical objects surrounding the analyzed system come from.

The irreversibility problem can neither be solved within the dynamical chaos theory, since the chaoc in question is a determined and reversible one.

The classical mechanics was so spectacularly successful and its predictions were so exact, that it didn't leave great Boltzmann the only and the decisive argument in the discussion with his opponents - he couldn't have made a statement about the classical mechanics theory not being complete.

Until early 20th centure there were no experimental data demonstrating the incompleteness of the classical mechanics. The study of black body radiation, atom spectra and photoeffect has lead to the creation of quantum theory. But with all the empressive achievements of quantum mechanics, there is our common sense and the fact, that the heat always goes from a hot object to a cold one, which prevent us from deeming the quantum mechanics absolutely exact. This makes it crusual to obtain experimental data which would not be describable on the basis of quantum mechanics and would demonstrate the manifestation of the 2nd law of thermodynamics at the evolution of macrosystems.

It is practically impossible to solve the mechanics equations for the macrosystems in which the thermodynamics laws apply. But if the mechanical state of the system cannot be described, how can we compare the predictions of mechanics with those of thermodynamics? Nonetheless, such comparison is possible, and it turns out that it does not require solving the mechanics equations. This comparison can be made on the basis of real experiments in changing the time sign in the equations which describe the macrosystem behaviour.

1.1 Possibility of time reversion experiments

It is possible to speak about a time-reversing experiment in principle based on the fact that changing the system Hamiltonian sign is equivalent to changing the time sign. This statement becomes obvious if we present the system wave function as

$$\Psi(t) = e^{-i\mathcal{H}t}\Psi(0) \tag{1}$$

or write down the Liouville equation solution for the system density matrix:

$$\sigma(t) = \exp(-i\mathcal{H}t)\sigma(0)\exp(i\mathcal{H}t) \tag{2}$$

We should bear in mind, though, that we are interested in the internal evolution of the system, which is determind by the interactions between all its particles. It might seem impossible to change the sign of interactions in macrosystems where the thermodynamics laws apply. Nevertheless, at least for spin systems, it turned out to be possible not only to change the Hamiltonian sign, but also to compare the forecasts of mechanics and the effects of the 2nd law of thermodynamics under those conditions.

The Hamiltonian of a spin system placed in a strong external constant magnetic field $H_0 = \frac{\omega_0}{\gamma}$, in a frame rotating with a frequency ω_0 around the axis Z reads [1]:

$$\mathcal{H} = \mathcal{H}'_d = \sum_{i<j} a_{ij}[I_{zi}I_{zj} - \frac{1}{4}(I_{+i}I_{-j} + I_{-i}I_{+j})] \tag{3}$$

If an alternating field of a resonant frequence and an amplitude ω_1/γ is applied to the system, then in the rotating frame we get:

$$\mathcal{H}_R = \omega_1 I_x + \mathcal{H}'_d \tag{4}$$

Going into the tilted rotating frame by a unitary transformation determined by the operator $\exp(-i\frac{\pi}{2}I_y)$, we obtain:

$$\begin{gathered} \mathcal{H}_{TR} = \omega_1 I_z - \tfrac{1}{2}\mathcal{H}'_d + \tfrac{3}{8}P, \\ P = \mathcal{H}_d^{(2)} + \mathcal{H}_d^{(-2)}, \quad \mathcal{H}_d^{(2)} = \mathcal{H}_d^{(-2)*} = \textstyle\sum_{i<j} a_{ij}I_{+i}I_{+j}. \end{gathered} \tag{5}$$

According to (2), we write down for the density matrix of the system with the Hamiltonian (5):

$$\sigma(t) = \exp\{-i(\omega_1 I_z - \frac{1}{2}\mathcal{H}'_d + \frac{3}{8}P)\}\sigma(0)\exp\{i(\omega_1 I_z - \frac{1}{2}\mathcal{H}'_d + \frac{3}{8}P)\} \qquad (6)$$

In a strong alternating field H_1, when $\omega_1 \gg \omega_L$ ($\omega_L = \gamma H_L$, where the local field $H_L = \{[Tr(\mathcal{H}'_d)^2]/[Tr(M_z^2)]\}^{1/2}$ [1]), the non-secular operator $3/8P$ in (6) can be neglected in the first approximation. Then, the internal evolution of the system will be described by the Hamiltonian $-\frac{1}{2}\mathcal{H}'_d$, while the evolution in a rotating frame with no alternating field is described by the Hamiltonian $\mathcal{H}'_d$.

The transfer to a tilted rotating frame is a formal operation and cannot actually influence the system evolution. At the same time, the influence of alternating field 90°_y -pulse on the system is described by the same operator that describes the transfer to a tilted frame. Thus, if we combine the sufficiently long application of a strong alternating field to the system with the application of short pulse, we can create a situation where the internal interactions sign in the system Hamiltonian will be changed with an accuracy determined by the correlation between ω_1 and ω_L

1.2 Magic echo

The density matrix

$$\sigma = 1 - \beta\omega_0 I_x \qquad (7)$$

describes the state of the of the spin system in a strong external constant field after the 90°_y-pulse was applied to the system. Here β^{-1} is the system temperature.

The transverse component of magnetization $I_x(t)$ causes the free induction signal.

The exciting experiments of Rhim, Pines and Waugh [2] have shown, that the free induction signal restores in time which is much longer than the spin-spin relaxation time T_2 (T_2 is defined as the time required for the free induction signal decay under normal conditions). Before the experiments [2] this time was considered as a thermodynamic relaxation time in spin systems. The phenomenon observed in [2] was called "magic echo".

The transverse component of magnetization operator $I_x(t)$, to which the free induction signal corresponds, is non-diagonal in the energy representation. Consequently, the 2nd law of thermodynamics should not apply to the $I_x(t)$ evolution. Meanwhile, if the irreversibility of macrosystems evolution does exist, and if we define the time of equilibrium state establishing in the system as the time of irreversible disappearance of the non-diagonal matrix elements in the density matrix, we require the condition

$$\frac{\partial \sigma}{\partial t} = -i[\mathcal{H}, \sigma] = 0 \tag{8}$$

to be fulfilled in equilibrium, which means that the density matrix should become diagonal in energy representation. The disappearance of the non-diagonal matrix elements in the density matrix at the irreversible process of establishing the equilibrium in the system should, logically, also be irreversible.

The fact that the $I_x(t)$ evolution was reversible in the experiment [(2)] made the authors come to a conclusion, that the spin temperature concept should be treated cautiously.

Thus, on the one hand, the spin temperature concept is confirmed by a huge number of experimental data, and it is hard to doubt that the spin system energy levels are populated according to the Boltzmann distribution. On the other hand, the non-diagonal operator $I_x(t)$ in the system density matrix does not irreversibly disappear during a time period which considerably surpasses T_2. If we assume that the spin temperature concept is correct, and that the spin system evolution is really irreversible, then the experiments [(2)] can lead us to a conclusion that T_2 is not a thermodynamic relaxation time.

If the $I_x(t)$ evolution in experiments [(2)] turned out to be irreversible, the authors of [(2)] would not have had a basis for claiming that the spin temperature concept should be treated cautiously. But on the other hand the $I_x(t)$ evolution in that case would not have been describable on the basis of mechanics approach. Such result would have directly demonstrated the incompleteness of the quantum theory and the significance of it would have been equivalent to this fact.

The operator $I_x = \sum_i I_{xi}$ has a simple structure and is a sum of one-particle operators. The modern pulse NMR has the methods [(1)] which allow

to bring the spin system to a state described by the density matrix

$$\sigma = 1 - \beta \mathcal{H}'_d \tag{9}$$

with high value of the inverse temperature β.

When θ_y-pulse is applied to the spin system, the density matrix is transformed to [1]:

$$\sigma_R = 1 - \beta[\frac{1}{2}(3\cos^2\theta - 1)\mathcal{H}'_d + \frac{3}{8}P\sin^2\theta - \frac{3}{4}Q\sin\theta\cos\theta] \tag{10}$$

where

$$Q = \sum_{i<j} a_{ij}[I_{zi}(I_{+j} + I_{-j}) + I_{zj}(I_{+i} + I_{-i})].$$

and P had been determined by (5).

Operators P and Q correspond to the interactions of all particles of the spin system. It would be natural to expect that the evolution of those operators would be different from the operator I_x evolution. The paper [3] studied the behaviour of the operator P and Q under the condition of the time sign change. In energy representation, Q has only non-diagonal matrix elements, but the energy reservoir corresponds to the operator $(3/8)P$ at the time of alternating field being applied. The experiments were carried out on the nuclei spin system of ^{19}F in a single crystal CaF_2. A dipole magic echo was discovered - the restoration of the signal confined to the dipole interactions in the time that considarably surpasses T_2. It turned out that the peculiarities in the behaviour of the operator $(3/8)P$ to which the energy reservoir corresponds cannot be explained on the basis of reversible equations of mechanics.

The purpose of paper [3] was to find the dipole magic echo and to compare the behaviours of the magic echo signal from P and Q operators. The measurements were performed in one crystal orientation with respect to the external constant magnetic field at a relatively small range of the alternating field amplitudes.

Very important conclusions regarding the irreversibility can be made from the results of [3]. That is why the need for the continuation of the time reversing experiments in spin systems is obvious. Besides, some aspects of [3] require correction.

2 Experiments, discussion

In this paper we observe and study the dipole magic echo in a wide range of external alternating field values at various crystal orientations relative to the constant magnetic field.

The measurements are performed on the CaF_2 single crystal, which is very convenient for this type of experiments. The avarage value of dipole-dipole interactions in different orientations is proportional to local field ω_L/γ, where ω_L is determined by $\omega_L = (M_2/3)^{1/2}$ [1]. Using the value of the second moment M_2 of the NMR line for CaF_2, given by Abragam [4] we find in the case of [111], [110] and [100] orientations ω_L/γ= 0.88, 1.25, 2.01 G respectively.

The pulse sequences which is similar to those presented in [3] were used to achieve the goals. The temperature was 300 K. The time of nuclei spin-lattice relaxation was 8 s. Since the duration of the time reversing part of the applied pulse sequences was not more than 1 ms, so the analized system may be considered isolated from the lattice during this time.

2.1 Evolution of the (3/8)P dipole subsystem under the conditions of time reversion

We studied the operator P evolution under the conditions of the time reversion by pulse sequence 1 (Fig.1).

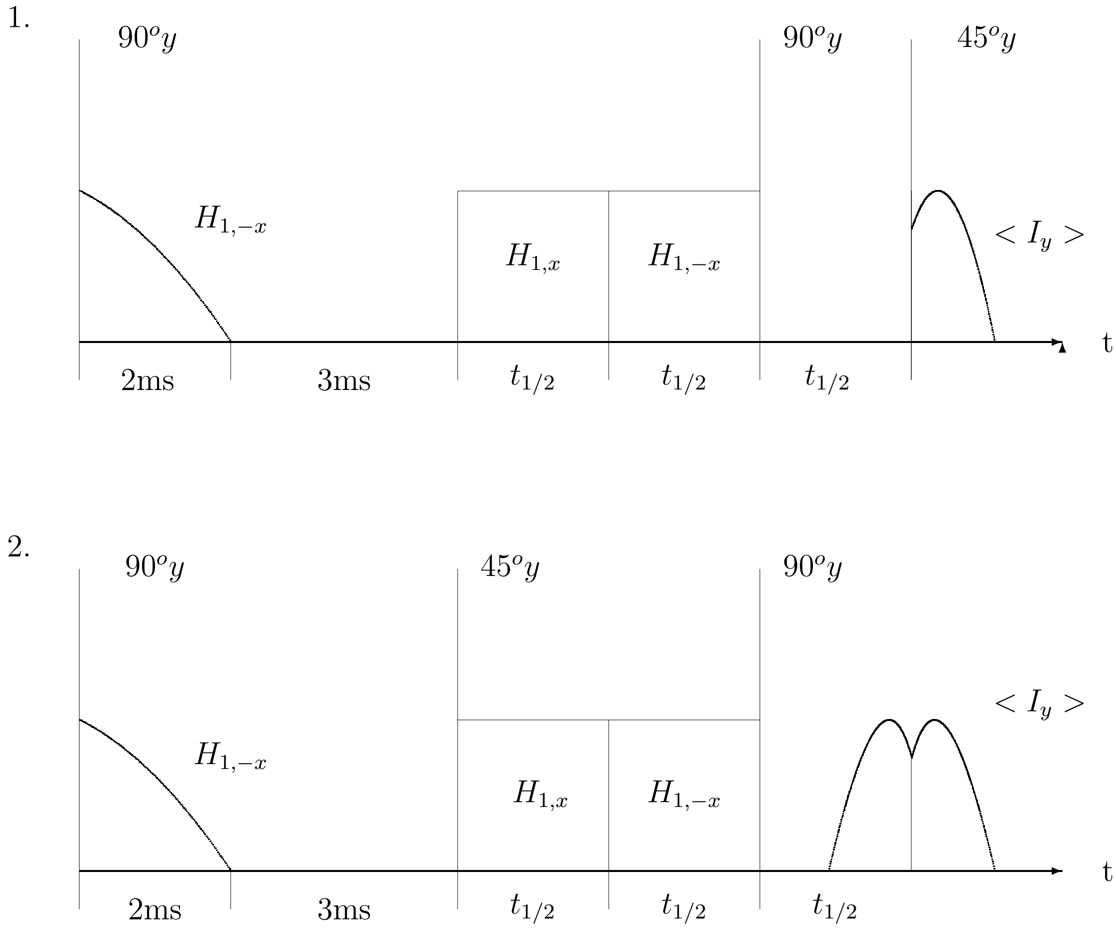

Fig.1. Pulse sequences used

The alternating field phase was changed to opposite one in the middle of the time interval of the application of this field. The adiobatic demagnetization in a rotating frame brings the system under consideration to a state which is described by the density matrix (9). The system Hamiltonian in a tilted rotating frame during the time of alternating field application is (5),

and the initial density matrix is

$$\sigma(0) = 1 - \beta(\frac{3}{8}P - \frac{1}{2}\mathcal{H}'_d) \qquad (11)$$

If the spin system obey the thermodynamic laws, it should be expected that after the equilibrium is established in the system the density matrix will look:

$$\sigma_{eq} = 1 - \beta_1(\omega_1 I_z + \frac{3}{8}P - \frac{1}{2}\mathcal{H}'_d) \qquad (12)$$

The experiments in laboratory [1] and a rotating [5] frame have demonstrated, that the unified spin system temperature is established in two stages. At the first stage, the Zeeman reservoir and the non-secular dipole-dipole interactions reservoir (to which the operator $(3/8)P$ corresponds in our case) undergo the quick warm mixing. At the second stage, the temperature of the newly created subsystem and of the reservoir of the secular part of the dipole-dipole interactions get levelled at a slower pace.

2.1.1 Description of the operator P evolution by reversible equations

Let's review the spin system evolution at the application of the pulse sequence 1 on the basis of expression (2). The transformation which corresponds to the 90°_y-pulse effect and to the transfer to the tilted rotating frame compensate each other. That's why by the time the 45°-pulse is applied we get:

$$\begin{aligned}
&\sigma(\frac{3}{2}t_1) = A_1\sigma(0)A_1^{-1} \\
&A_1 = \exp(-i\mathcal{H}'_d\frac{t_1}{2})A_-A_+ \\
&A_\pm = \exp\{-i(\pm\omega_1 I_z + \frac{3}{8}P - \frac{1}{2}\mathcal{H}'_d)\frac{t_1}{2}\} \qquad (13)
\end{aligned}$$

If the 45°-pulse is excluded from the pulse sequence 1, then after the alternating field is removed the spin system signal is absent. The signal observed after the 45°-pulse is determined by the transverse component I_y, and we can write $\langle I_y\rangle = \langle I_y\rangle_1 + \langle I_y\rangle_2$. The value $\langle I_y\rangle_2$ is the contribution to $\langle I_y\rangle$ which is bound to the density matrix operator $-\frac{1}{2}\mathcal{H}'_d$. The results of [5] demonstrate, that this contribution is constant under the conditions of our experiments and can be easily subtracted from the signal under observation.

If $\omega_1 \gg \omega_L$, then in the first approximation we can write

$$
\begin{aligned}
&P(t_1+t) = A_2 P A_2^{-1} \\
&A_2 = \exp\{-i\mathcal{H}_d'(t-\frac{1}{2}t_1)\}
\end{aligned}
\qquad (14)
$$

It is demonstrated in [3], that in the case of $t_1 = N\frac{\pi}{\omega_1}$ from (14) it follows:

$$
\langle I_y \rangle_1 = \frac{3}{16}\beta Tr(I_y^2)\frac{d}{dt}G(t) \qquad (15)
$$

where $G(t)$ determines the form of the free induction signal.

A signal whose amplitude does not depend upon the alternating field application time corresponds to (15).

Fig. 2 presents the dependence of the amplitude of the signal corresponding to the $(3/8)P$ operator in the density matrix upon the alternating field application time t_1.

It follows from the Fig. 2 that the signal in the pulse sequence 1 decays as t_1 grows. The signal decay turned out to grow with the transition from the [111] to [100] orientation, but not depend upon the ω_1 in every orientation.

If $t_1 = N\frac{\pi}{\omega_1}$, then, using the Magnus expansion [6], we find:

$$
\begin{aligned}
&\exp\{-i(\omega_1 I_z + \frac{3}{8}P - \frac{1}{2}\mathcal{H}_d')t\} = \exp(-i\omega_1 I_z t)\exp(-iFt), \qquad (16) \\
&F = -\frac{1}{2}\mathcal{H}_d' + \sum_{k=1}^{\infty}\frac{1}{(k+1)!}\frac{\mathcal{H}_k}{(2\omega_1)^k}
\end{aligned}
$$

The $\mathcal{H}_k$ operators have the dipole-dipole interactions magnitude in the $k+1$ power. The correction to the average Hamiltonian $-\frac{1}{2}\mathcal{H}_d'$, corresponding to the first term of the sum by k in (16), is

$$
\begin{aligned}
&\mathcal{H}^{(1)} = \mathcal{H}_1^{(1)} + \mathcal{H}_2^{(1)} \qquad (17) \\
&\mathcal{H}_1^{(1)} = (\frac{3}{8})^2\frac{[\mathcal{H}_d^{(-2)}, \mathcal{H}_d^{(2)}]}{2\omega_1} \quad \mathcal{H}_2^{(1)} = \frac{3}{8}\frac{1}{2}\frac{[\mathcal{H}_d', \mathcal{H}_d^{(-2)} - \mathcal{H}_d^{(2)}]}{2\omega_1}
\end{aligned}
$$

We shall not need an explicit form of the corrections to $-\frac{1}{2}\mathcal{H}_d'$ of a higher order.

If the alternating field phase changes often, like in [2], the odd power corrections to $-\frac{1}{2}\mathcal{H}_d'$ in (16) vanish [2,6]. In our experiments, the correction $\mathcal{H}^{(1)}$

does not disappear, but the signal decay contribution due to $\mathcal{H}^{(1)}$ decreases because of the phase change of alternating field.

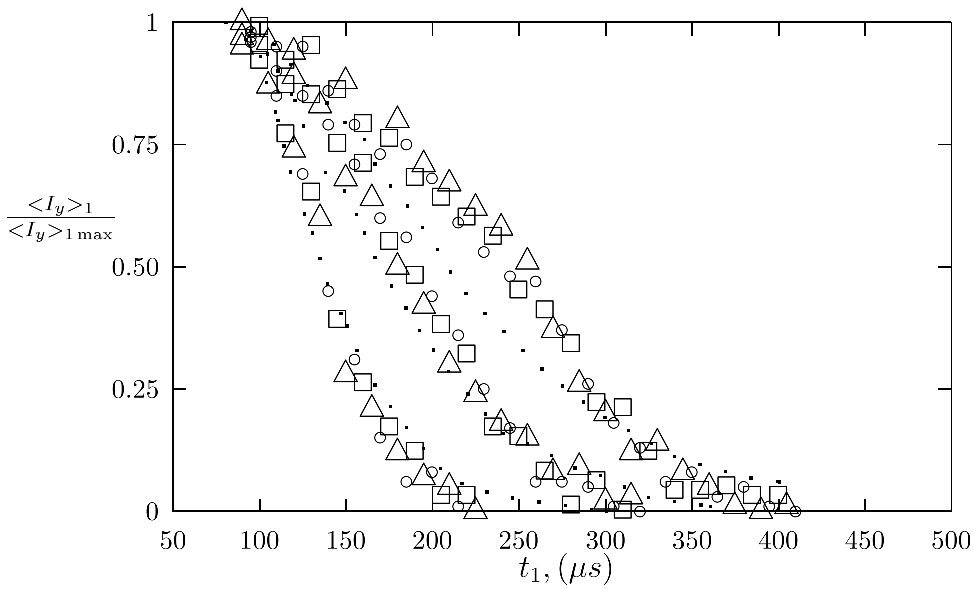

Fig.2. Dependence on t_1 of the amplitude of the magic echo signal due to the operator P at $\frac{\omega_1}{\gamma} = 12.5G(\square)$; $\frac{\omega_1}{\gamma} = 25.3G(\circ)$; $\frac{\omega_1}{\gamma} = 57.2G(\triangle)$ in the [100], [110] and [111] orientations

Using (16), by the time the 45°-pulse is applied we get (in the case of constant alternating field phase):

$$P(\frac{3}{2}t_1) = A_3 P A_3^{-1} \tag{18}$$
$$A_3 = T\exp[-i\int_0^{t_1}\exp(-\frac{i}{2}\mathcal{H}'_d t)\mathcal{H}_1^{(1)}\cdot\exp(\frac{i}{2}\mathcal{H}'_d t)dt]$$

Writing down (18) we neglected the terms with $k > 1$ in the expression for F (16), because under the conditions of our experiments the ratio of $-\frac{1}{2}\mathcal{H}'_d$ to the $\frac{1}{(k+1)!}\frac{\mathcal{H}_k}{(2\omega_1)^k}$ is of the order of 10^4 already for $k = 2$. Hence, the correction terms with $k > 1$ cannot contribute considerably to the decay

of the signal which is due to the operator $(3/8)P$. We ommitted also the non-secular operator $\mathcal{H}_2^{(1)}$ in expression for $\mathcal{H}^{(1)}$.

Taking into account the change of the alternating field phase we have:

$$P(\frac{3}{2}t_1) = A_4 P A_4^{-1} \tag{19}$$
$$A_4 = \exp(-i\mathcal{H}'_d \frac{t_1}{2}) \exp[i(\frac{\mathcal{H}'_d}{2} + \mathcal{H}_1^{(1)})\frac{t_1}{2}] \exp[i(\frac{\mathcal{H}'_d}{2} - \mathcal{H}_1^{(1)})\frac{t_1}{2}].$$

2.1.2 Analysis of the alternating field ungomogeneity influence on the experiments results

It was shown experimentally in [3], that the signal in the pulse sequence 1 decays much faster in the case of the constant alternating field phase. It would be natural to give the following explanation: when the alternating field phase is constant, the contribution to the decay grows by means of the operator $\mathcal{H}_1^{(1)}$. Besides, the changing of the alternating field phase reverses the direction of the isochromates precession in the rotating frame and compensates the field inhomogeneity influence on the signal under observation.

The time of the alternating field application in both phases was divisible by $\frac{\pi}{\omega_1}$. Fig. 2 demonstrates, that the signal decay at pulse sequence 1 does not depend upon the alternating field amplitude. At the same time, the signal decay time turned out to vary at different crystal orientation.

If it was operator $\mathcal{H}_1^{(1)}$ playing the major role in the signal decay in pulse sequence 1, then the signal would have been decaying at a slower pace when the ω_1 would grow. Also, if it was the not totally compensated field inhomogeneity that determined the signal dependence on t_1, the signal in pulse sequence 1 would have been decreasing when ω_1 would grow. Since the field inhomogeneity contribution to the signal decay is directly proportional to ω_1, and the operator $\mathcal{H}_1^{(1)}$ contribution is in reverse proportion to ω_1, it is also not possible to explain the signal independence from ω_1 by sum of those two contributions.

We have also achieved purely experimental evidence of the fact that the signal decay in pulse sequence 1 cannot be connected to the alternating field inhomogeneity. Indeed, the alternating field inhomogeneity does not depend upon the crystal orientation, and the fact that the signal heavily depends on the orientation points out that the signal decay is not determined by the field inhomogeneity even at its maximum. Finally, we have taken the

measurements where we used the coils of varying size to create the alternating field, which, obviously, produced the fields of different homogeneity. However, the measurement results did not change when the coils were changed. It would also be mentioned that the measurements were performed in this work and in [3] using different equipment and different samples. The results for the same orientations and with the same ω_1 value coinsided, which is the evidence of there correctness.

2.1.3 Irreversibility of the $(3/8)P$ subsystem evolution

The pulse sequence "c" used in [3] allow to measure the decay of $(3/8)P$ operator signal, which is due to the operator $-\frac{1}{2}\mathcal{H}'_d$ in the Hamiltonian (5). This decay time occurs in orientation [111] was equal to 120 μs.

The operator $\mathcal{H}_1^{(1)}$, which, if the quantum mechanics description is correct, determines the dipole magic echo signal decay, is on the two orders of magnitude smaller than the operator $-\frac{1}{2}\mathcal{H}'_d$, when the values of ω_1 are those used in our experiments. Correspondingly, even not taking the alternating field phase change into account, the signal decay time t_d in pulse sequence 1 should be of two orders longer than 120 μs. The alternating field phase change reverses the sign of ω_1 and this increases the expected time t_d even more (see (18) and (19)). Thus, there is ground to think, that the decay times t_d which are observed at pulse sequence 1 and do not exceed 350 μs are much shorter than the decay time that follows from the system description based on expressions (18) and (19) which follows from (2).

Next, at the transfer from one orientation to another, the value of the operator $\mathcal{H}_1^{(1)}$ changes in proportion to the second power of the local field, i.e. in proportion to the change of the second moment M_2, which grows 5 times when the system transfered from orientation [111] to orientation [100] [4] . Hence, the difference between the value of the operator $\mathcal{H}_1^{(1)}$ in the orientation [100] when $\frac{\omega_1}{\gamma} = 12.5$G and in the orientation [111] when $\frac{\omega_1}{\gamma} = 52.6$G is 20 times, and the signal disappearence time should also be on 20 times different. Fig.2 demonstrates, though, that the times under comparison for the signal in pulse sequence 1 are not more than 2 times different.

Finally, the signal in the pulse sequence 1 does not depend on ω_1 contrary to the predictions of the theory based on equation (2).

Thus, the behaviour of the dipole-dipole interaction subsystem, to which the operator $(3/8)P$ corresponds, under the time reversion conditions cannot

be described on the basis of expressions (1) and (2). This fact leads us to the following coupled conclusions:
a) the evolution of the subsystem corresponding to the operator $(3/8)P$ in density matrix is irreversible;
b) the process of a system coming to an equilibrium state really is irreversible and cannot be described by the reversible expressions (1) and (2);
c) the macrosystem under analysis being isolated does not lead to the reversibility of its evolution.

At the same time, if the subsystem $(3/8)P$ evolution were reversible, it would been the evidence either of the fact, that the spin temperature concept is inapplicable, or of the fact that the system‘s thermodynamic relaxation time is much longer than the time of experiment, or of the fact that the mechanics equations are fantastically exact and the thermodynamics irreversibility is illusionary. But the 2nd law of thermodynamics is the generalization of the experience and the discussion of its correctness makes no sence, while the exactness of system evolution description on the basis of mechanics equations is limited by the extent of the mechanical theory completeness. The experimental results which we recieved studying the evolution of the subsystem of non-secular dipole-dipole interactions, to which the operator $(3/8)P$ corresponds, cannot be described by the reversible mechanics equations and, hence, demonstrate the incompleteness of those equations.

2.1.4 Thermodynamical description of the evolution of the $(3/8)P$ subsystem

We select the irreversible component of the evolution of the $(3/8)P$ subsystem using pulse sequence 1. It makes sence to introduce its temperature $\beta^{-1}(t)$ for the thermodynamical description of this subsystem irreversible evolution. The following integro-differential equation for the inverse temperature $\beta(t)$ was obtained in [7] on the basis of non-equilibrium thermodynamics methods:

$$d\beta/dt = -\int_0^t \beta(t')G_1(t'-t)dt', \tag{20}$$

where

$$\begin{aligned} G_1(t'-t) \;=\; & \tfrac{3^2}{8^2\langle\mathcal{H}_d^{(2)}\mathcal{H}_d^{(-2)}\rangle}\textstyle\sum_{i>j}\langle[\mathcal{H}_{dij}^{(2)},\mathcal{H}_d^{(-2)}]\exp(i\mathcal{H}'_d(t'-t)/2)\times \\ & [\mathcal{H}_{dij}^{(-2)},\mathcal{H}_d^{(2)}]\exp(-i\mathcal{H}'_d(t'-t)/2)\rangle \end{aligned}$$

It can be seen from (20), that the rate of the reverse temperature change does not depend on ω_1, which corresponds to the fact that the signal observed in pulse sequence 1 does not depend on ω_1. The $G_1(t)$ function cannot be calculated explicity. Let's write it down as

$$G_1(t'-t) = (n\omega_L)^2 G_2(t'-t) \tag{21}$$

where the value of n is of the order of 1. Let us use for $G_2(t)$ the Gaussian approximation, which is natural for regular magnetic of the CaF_2 type [7]:

$$G_2(t'-t) = \exp(-\frac{1}{2}M(t'-t)^2). \tag{22}$$

The value M must be comparable to the second moment M_2 of the NMR line in CaF_2. We get $M_2 = 2.55 \cdot 10^{10}$, $0.99 \cdot 10^{10}$ and $0.5 \cdot 10^{10}$ s^{-2} for the [100], [110] and [111] orientations [4]. Since operator $-\frac{1}{2}\mathcal{H}'_d$ is in the function $G_1(t)$ exponent, let's write $M = M_2/4$. The dotted lines in the Fig.2 correspond to the equation (20) solution when n = 0.45 and when $M = M_2/4$ for every orientation. The agreement with the experiment is the best when the starting point is moved 80 μs in t in the equation (20). It can be explained by the fact, that when time t_1 is less than 80 μs, the system stochastization after the alternating field application does not show yet. Anyhow, there was no considerable signal decay at t_1 less than 80 μs.

A total coincidence of theory and experiment when the relaxation processes are described by the non-equilibrium thermodynamics methods seems somewhat suspicious. That is why we decided not to try and select such version of the correlation function $G_1(t)$ which would make the calculated curves to reproduce the experimental data exactly. Besides, the change in the alternating field phase can influence the subsystem $(3/8)P$ thermodynamics evolution and introduce changes in the equation (20) description of this evolution, especially at small values of t_1.

It can be seen from Fig. 2, that in the case of Gaussian approximation of $G_2(t)$ function, when the parameter values correspond to the sample under observation, the eguation (20) solution describe the character of the signal dependence on orientation and time very well. The degree of the quantitative correspondence of the theory [7] to the measurement results may be considered quite enough.

2.2 Evolution of non-diagonal operator Q under the condition of time reversion

Let us now review the spin system evolution at pulse sequence 2 (Fig.1). After the 45°-pulse the system state is described by the density matrix

$$\sigma = 1 - \frac{1}{4}\beta(\mathcal{H}'_d + \frac{3}{4}P - \frac{3}{2}Q) \tag{23}$$

The operator Q in (23) causes the signal [(1)]:

$$\langle I_y \rangle = \frac{3}{8}\beta Tr(I_y^2)\frac{d}{dt}G(t) \tag{24}$$

Similar to (19) we obtain:

$$Q(\frac{3}{2}t_1) = A_4 Q A_4^{-1}. \tag{25}$$

In accordance with (25), when $t = \frac{3}{2}t_1$, a signal confined with the operator Q appears without any additional influence on the system. The amplitude of this signal turned out to depend upon both the orientation of the sample and the alternating field amplitude. Hence, the measurement results are presented on Fig.3 - Fig.5. Figs. 3 and 4 demonstrate the dependence of the dipole magic echo signal upon the alternating field application time in the orientation [100] and [110] when the ratio $\frac{\omega_1}{\gamma}$ was changed in the interval 12.5 - 52.7 G.

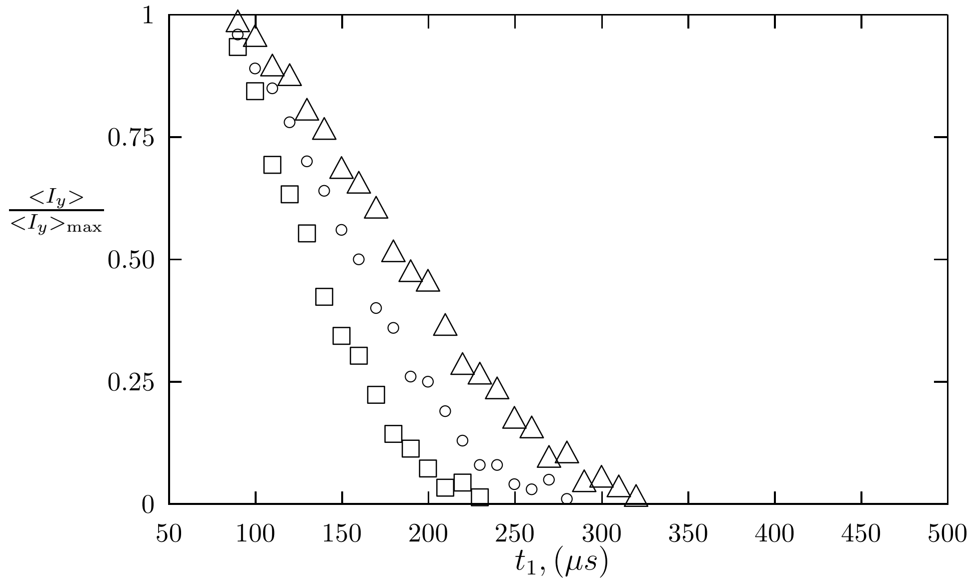

Fig.3. Dependence on t_1 of the amplitude of the magic echo signal due to the operator Q at $\frac{\omega_1}{\gamma} = 12.5G(\square)$; $\frac{\omega_1}{\gamma} = 25.3G(\circ)$; $\frac{\omega_1}{\gamma} = 52.7G(\triangle)$ in the[100] orientation

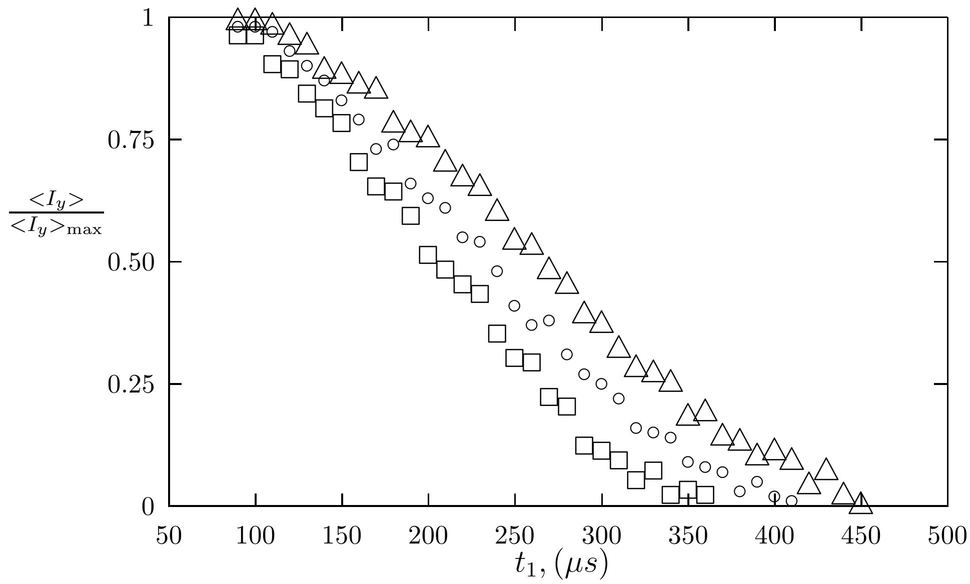

Fig.4. Dpendence on t_1 of the amplitude of the magic echo signal due to the operator Q at $\frac{\omega_1}{\gamma} = 12.5G(\Box)$; $\frac{\omega_1}{\gamma} = 25.3G(\circ)$; $\frac{\omega_1}{\gamma} = 52.7G(\triangle)$ in the [110] orientation

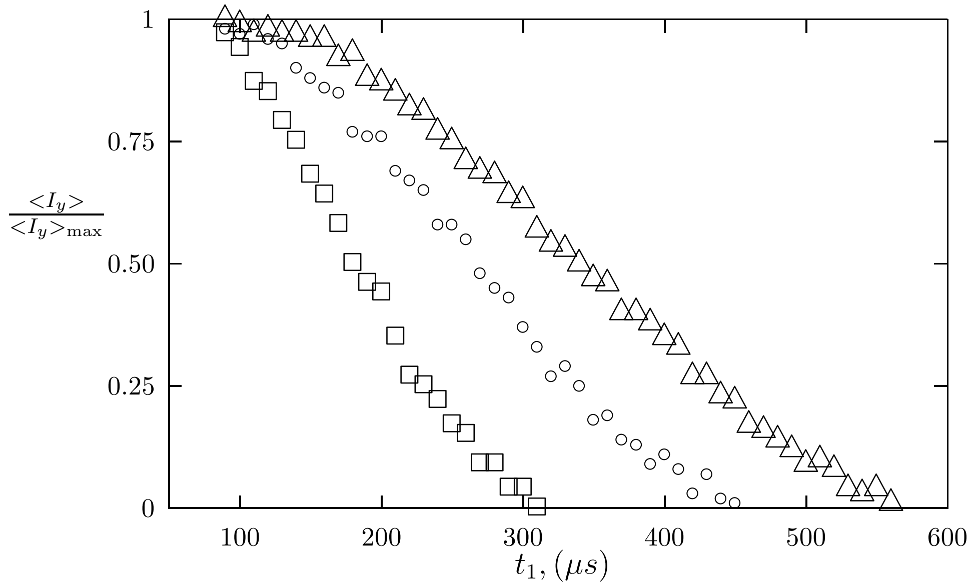

Fig.5. Dependence on t_1 of the amplitude of the magic echo signal due to the operator Q at $\frac{\omega_1}{\gamma} = 52.7G$ in the [100] - (□); [110] - (○); [111] - (△) orientation

Fig.5 demonstrates the signal dependence on t_1 with $\frac{\omega_1}{\gamma} = 52.7$ G at various crystal orientations. The measurement results for orientation [111] at $\frac{\omega_1}{\gamma}= 12.5$ G and $\frac{\omega_1}{\gamma} = 25.3$ G coinside with those in [3] and thus are not presented here.

The facts that the signal in pulse sequence 2 grows when ω_1 grows and that the signal depends on orientation demonstrate that the alternating field inhomogeneity influence on the effects under observation is neglectable small. If we consider that it is the operator $\mathcal{H}_1^{(1)}$ that causes the decay of the signal in pulse sequence 2, then the amplitude should grow when P grows, and decrease when ω_1 grows. Figs. 3-5 show, that the experimental data qualitatively correspond to this conclusion. The inference that the operator Q evolution is reversible when a time-reversing pulse sequence is applied to the system was made in [3], where the measurements were taken only in one orientation, based on the signal dependence on ω_1. However, in present work we have analized the whole set of experimental data that we obtained when studying the Q magic echo in various orientation and with a wide range of

ω_1 values, and this analysis questions the correctness of above conclusion.

Indeed, the signal decay time in pulse sequence 2, similar to the pulse sequence 1, proves to be much less than that corresponding to presence of the operator $\mathcal{H}_1^{(1)}$ in the expression for A_4. Besides, the decay times in orientation [111] at $\frac{\omega_1}{\gamma} = 52.7$ G and in orientation [100] at $\frac{\omega_1}{\gamma} = 12.5$ G are not more than 3 times different, while the value of the operator $\mathcal{H}_1^{(1)}$ on 20 times differs. Thus, it is difficult to reconsile the idea proposed in [3] of the operator Q evolution being reversible at pulse sequence 2 with the results of our experiments.

The signal decay time t_d in pulse sequence 2 is longer than that in pulse sequence 1, but values of t_d remain in the same order of magnitude. According to the basic statements of statistical physics, the evolution results for the diagonal terms of the density matrix and the non-diagonal ones are totally different. Hence, the characteristics of the P and Q operators evolution in the equilibrium establishing process should also be different. In our experiments, this difference appears in the fact that the signal corresponding to the operator Q depends on ω_1.

2.3 Comparison of the peculiarities of free induction signal magic echo and dipole magic echo

In ref.[2], the alternating field phase was changed every $\frac{\pi}{\omega}$seconds when the time reversion situation was created. As a result, the corrections to the mean Hamiltonian introduced by the odd powers of ω_1 become a zero, and the alternating field inhomogeneity gets compensated well. The noticable free induction signal magic echo was observed up to $t_1 = 650$ μs. The authors in [2] see the reason for the signal decay in the influence of the alternating field phase-changing pulses non-idealities. Actually, the alternating field amplitude in [2] was 100 G, and 500 pulses correspond to 650 μs of the alternating field application. At that pulse rate there really is ground to assume that the magic echo signal decay is explained by the pulse non-idealities.

We can not say how a very frequent alternating field phase change influences the irreversible component of the system evolution. But the possible demonstrations of the system evolution irreversibility under the physical conditions which are created by the phase changing field, can be different from the case of the long alternating field application without a phase change. It is

therefore possible, that the time of the feasible irreversibility demonstration when a phase-changing field influences the system exeeds the signal decay time as a result of the pulse non-idealities.

In our experiments the pulse non-idealities could not have influenced the observed signals considerably. But the time of the dipole magic echo signal decay turned out to be of the same order of magnitude as the time of the free induction signal magic echo decay. It gives us ground to assume, that the evolution irreversibility of the transverse component of magnetization I_x can contribute to the signal decay in the experiment [(2)].

On the other hand, in our experiments the energy redistribution occurs in spin macrosystem both in pulse sequence 1 and pulse sequence 2. This redistribution, in accordance with 2nd law of thermodynamics, is irreversible and the irreversible change of density matrix diagonal terms corresponds to this redistribution.

The operators, giving diagonal and non-diagonal density matrix terms, are expressed by means of the same one-particle operators of nuclear moment components. Correspondingly, the irreversibility of the evolution of the density matrix diagonal terms may lead to the irreversibility of the non-diagonal terms evolution also.

At the same time, in conditions of the experiments [(2)], the energy redistribution does not occur in spin macrosystem. As a result, the character of the operator Q and I_x evolution in time reversion experiments may be different.

We would like also to stress here, that operators P and Q are many-particle ones and their evolution may be distinguished essentially from the evolution of operator I_x because of this reason.

We see that the comparison of the paper [(2)] and our experiments results give rise to many questions. The answers on this questions may be given by new experiments only.

3 Conclusion

The study of the dipole magic echo that we have performed in a wide range of the values of ω_1 and at various crystal orientations, allow us to make the following main conclusions:

1) It turned out to be impossible to describe the evolution of the $(3/8)P$ subsystem in the time reversion situation on the basis of expressions (1) and

(2). The process of the system transfer to the equilibrium state, which is described by the density matrix (12) in the tilted rotating frame, is irreversible.

2) The dependence of the dipole magic echo signal confined with the non-diagonal operator Q upon ω_1 and upon the value of the dipole-dipole interactions corresponds qualitative to formula (25) which is derived from (2). But the quantitative evaluations demonstrate, that the signal decay in pulse sequence 2 happens much faster than it follows from the description based on the reversible expresions (1) and (2).

Probability assumptions are used in a apparent or hidden form when a transfer from the reversible mechanics equations to the irreversible non-equilibrium thermodynamic ones takes place. As a role, the introduction of those assumptions is explained by the facts, that the mechanics equations can not be solved exactly and that it is necessary to turn to the shortened description of the non-equilibrium processes in the macroscopic systems. The experimental and theoretical research performed in [(3)], [(7)] and in this paper demonstrate, that it is not the matter of the impossibility of solving the reversible mechanics equations exactly, but the matter of their inapplicability to the non-equilibrium processes in the macrosystems which obey the 2nd low of thermodynamics. This circumstance will allow to take a new view at the problem of chaos, both the classical and quantum ones.

The research that we have performed by far does not exhaust the unique possibilities which the dipole magic echo phenomenon presents for the study of the correlation between the reversibility and irreversibility in the macrosystems evolution. We believe that the continuation of the time reversing experiments in the spin systems will bring a new and, possibly, unexpected results.

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